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All References should be given in the text by quoting the surname of the author, followed by year of publication, *e.g.*, (Roy, 1958). The full Reference should be given in a list at the end, arranged alphabetically, as follows; Roy, S. B., 1958, *Ind. J. Phys.*, **32**, 323.
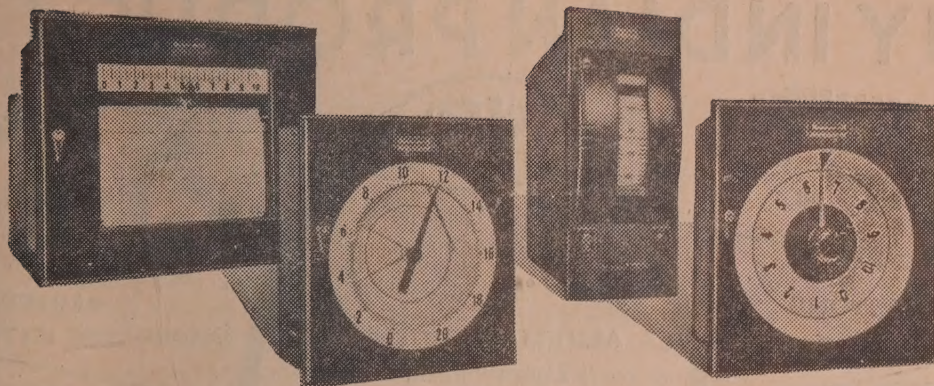
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$$A \cdot B \cdot E \cdot F \cdot G \cdot M \cdot P \cdot T \cdot W \cdot$$

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The mathematical expressions should be written carefully by hand. Care should be taken to distinguish between capital and small letters and superscripts and subscripts. Repetition of a complex expression should be avoid by representing it by a symbol. Greek letters and unusual symbols should be identified in the margin. Fractional exponents should be used instead of root signs.

TRANSISTOR DRIVE CIRCUITS FOR DEKATRONS

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ABSTRACT. Transistor drive circuits have been described for GC10/4B dekatrons by Chaplin and Kandiah (1958). These apply paired pulses of 80 μ sec duration and 80 volts amplitude to the dekatron guides and claim a resolving time of 300 microseconds.

A transistor circuit suitable for the faster GC 10D dekatron that attains a resolving time of 40 μ sec has been described by the authors. This supplies a 20 μ sec, 150 volt pulse and is essentially a blocking oscillator pulser using a ferrite core transformer. The transformer design details have also been described. The circuit is simple and reliable and works over a supply voltage range of 7.5 volts to 18 volts. The current drain at pulse rate of 5000 is only 2.0 mA at 12 volts. This is strikingly low when one remembers that the current drain for a paired pulse drive circuit is 10 mA at a pulse rate of 500 only.

Following this, a similar circuit for GC 10B, that needs 60 μ sec pulses was developed. At 5000 pulse rate its current drain is 12 mA at 12 volts supply. This circuit also works over a range of 7.5 volts to 18 volts. It is essentially more simple and reliable compared to the paired pulse drive circuits and shows a resolving time (200 μ sec) that is better than the manufacturers figure for this dekatron.

INTRODUCTION

Of all devices used for decade counting, the glow transfer type dekatrons appear to be the most simple. Extremely low operating power (0.15 watts) and reliability are its further attractions. Combined with a transistor drive circuit and transistorized high tension and extra high tension, perhaps the last word is reached in the development of a portable nuclear counting system that is simple and reliable, efficient and elegant.

Transistor drive circuits have been described by Chaplin and Kandiah (1958). These are for use with the neon-filled GC 10/4B dekatrons that have 250 μ sec resolving time. Paired pulses of 80 μ sec duration and 80 volts amplitude with adequate overlap, are applied by these circuits to the two guide electrodes of these dekratons. Resolving time of 300 μ sec has been claimed for these circuits, which use five crystal diodes, five resistors, two capacitors and a special transformer using a ferrite core. These circuits are adequate for nuclear counting system using G.M. Counters that have dead times in the range of 100 microseconds.

The scintillation detector, which gives a very great improvement in detection efficiency for γ-rays, is however capable of much faster counting. Scintillation

γ-ray spectrometers commonly utilize a pulse width of one microsecond. Very high counting rates (a million per minute) are often encountered. Counting systems must therefore have resolving times of one to five microseconds.

Such short resolving times are offered by transistor binary scaling circuits (Chaplin, 1956). We have found them equally reliable but the power consumption, components and cost per scale of two, exceed that for the drive circuit of a dekatron.

An argon filled dekatron is also available that has a resolving time of 50 μ sec. It is the three guide single pulse GC 10D that requires a 145 volt, 25 μ sec driving pulse. A transistor drive circuit for this tube is a more difficult design problem, chiefly because of the bigger pulse amplitude required with the shorter duration. Besides, since the preceding binaries will smooth out the randomness in the arrival of pulses to it, average counting rates to which this circuit may be subjected may run to 10,000 counts/sec. With such a high rate, power dissipation* in the driving transistor might be a cause for worry. Collector dissipation in common transistor types like OC 72 must not exceed 40 milliwatts for reliable operation in this country.

Fig. 1. GC 10D drive circuit

However, we have found such a circuit (Fig. 1) feasible. The most important component is evidently the ferrite core transformer. It utilizes interleaved windings as shown in Fig. 2. The insulation between windings is silicone tape and it was impregnated with Z 920 silicone varnish. Ferroxcube pot cores§ with the minimum unavoidable air gap, helps to keep down collector overshoot. The core does not saturate in the 20 μ sec pulse duration which is controlled by choosing the value of C (0.03 mfd) suitably. The circuit works over a supply voltage range of 7.5** volts to 18 volts. If the base bias is reduced, the circuit would work up to even 4.5 volts, but the repetition rate must not exceed 500 per

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*The average current in one of our units for GC 10 B, using a paired pulse drive circuit of Kandiah type, exceeds 10 mA with a repetition rate of only 500 per second.

§Philips D-36/22-10,00 III B1

**It is surprising to find that the GC 10 D operates reliably over a pulse amplitude range of 100 to 250 volts, in this circuit, whereas the manufacturer recommends only 144+50 volts −12 drive pulses. This has been verified in respect of all the twenty tubes we had in stock.

second, and the trigger pulse width must be more than 5 μ sec. This limitation in the repetition rate is probably due to the increase in the average $d-c$ level of the drive pulses at higher pulse rate so that higher amplitude drive pulses will be needed for a glow transfer. Also for a constant value of C, reduction in the supply voltage necessitates a longer duration trigger pulse for maintaining regeneration in the circuit. The current drain at 12 volts is only 2 mA with a pulse rate of 5000 per second. The trigger pulse need have a width of 2 microseconds and a little over 1.5 volts* in amplitude. Such a pulse is readily obtained from the preceding transistor binary.
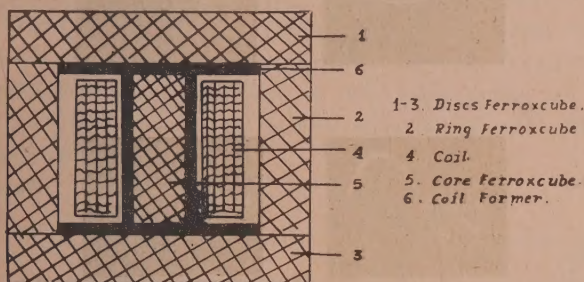

1-3. Discs Ferroxcube.
2. Ring Ferroxcube
4. Coil.
5. Core Ferroxcube.
6. Coil Former.

Fig. 2(a). Design of the blocking oscillator transformer.

B + E + H = Primary Winding (GC10D)
Q = ,, ,, (GC10B)
C + F = Secondary Winding (GC10D)
P + S = ,, ,, (GC10B)
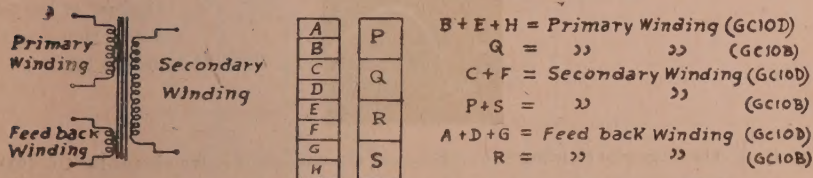A + D + G = Feed back Winding (GC10D)
R = ,, ,, (GC10B)

Fig. 2(b). Details of the coil winding of transformer in the
GC10D and GC 10B circuits.

The resolving time (and other performance data reported here) was measured in the arrangement of Fig. 3 and was found better than 50 microseconds. The

A = Fixed position Pulse.
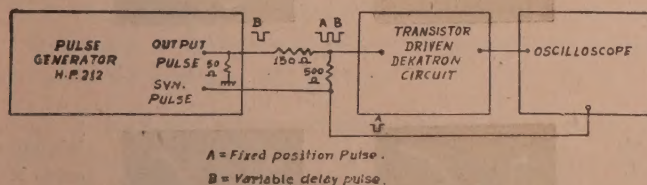B = Variable delay pulse.

Fig. 3. Arrangement for the measurement of the resolving time of the circuits.

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*The amplitude of the trigger pulse should be a little greater than the base bias. Smaller bias makes the circuit more sensitive.

oscillograms may be seen in Fig. 4a.   The large overshoot that follows each drive pulse in it (and also in Fig. 4b) rules out the possibility of glow resting on any of the transfer electrodes.   Furthermore, an appreciable overlap is automatically maintained in these circuits (Fig. 1 and Fig. 5).   Overlap ensures smooth transfer of the glow from the cathode to the first, second and third guides successively and adds to the inherent reliability of the operation of the tubes.

Waveform at the collector

Waveform at the guide No. 1.

Waveform at the guide No. 2.

Waveform at the guide No. 3.

Output of the transistor drive circuit for GC 10D
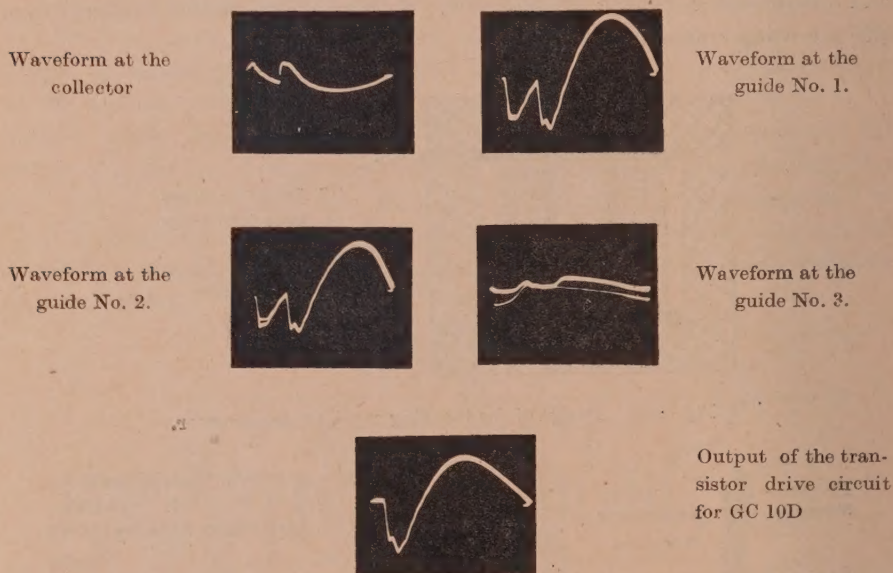
Fig. 4(a).   Oscillograms showing the response of the transistor drive circuit for GC 10D, to two input pulses separated by an interval of about 50μ sec.

Waveform at the collector

Output of the transistor drive circuit for GC 10B.

Waveform at guide No. 1.
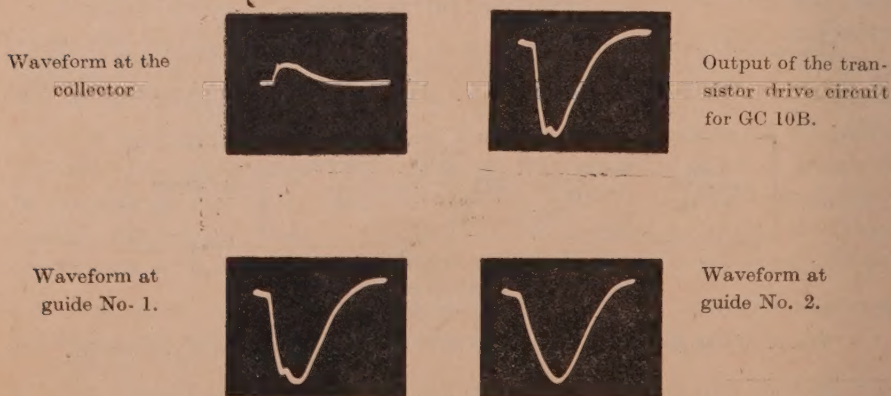
Waveform at guide No. 2.

Fig. 4(b).   Oscillograms showing the response of the transistor drive circuit for GC 10B.

The surprisingly low current drain of this circuit prompted an enquiry if a circuit for GC 10 B could be developed on these lines which would have much lower current drain. It was felt that the larger current drain of the paired pulse drive circuits (Chaplin, Kandiah) was due to two causes. First, one has to supply the power for storing adequate energy in the magnetic fields of the core, so that a large overshoot, necessary for generation of the second pulse, is produced. Secondly, one has to supply the power dissipated in the back resistance of the crystal diodes that switch the pulses on the dekatron guide electrodes. It was felt that an integrated pulse drive circuit (Ericsson Handbook) instead of the paired pulse, would have greatly reduced current drain. The circuit, as finally developed, is shown in Fig. 5. The resolving time has not been sacrificed in the least, in as much as the circuit counts correctly up to 5000 pulses per second, whereas the manufacturers specification is only 4000 pulses per second. The current drain at 5000 pulse rate is only 12mA at 12 volts which represents tremendous improvement.
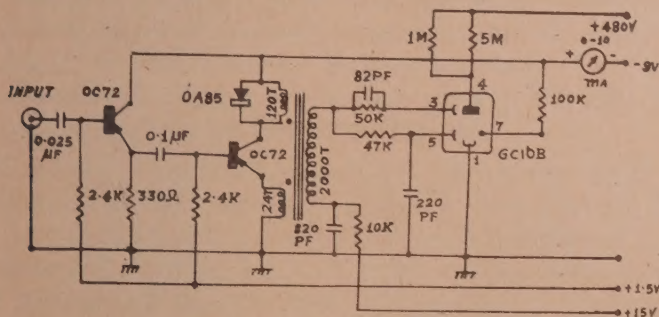

Fig. 5. GC 10B drive circuit.

The circuit works reliably over a supply voltage range of 7.5 volts to 18 volts and it will be seen (Fig. 5) that it is simpler than the paired pulse circuit. In the paired pulse circuits of Chaplin and Kandiah, high back voltages to which the switching diodes across dekatron guides are subjected, introduce a likely cause of failure. Reliability is further endangered in these circuits as the transistor is also subjected to the extremes of its ratings.

The design of the ferrite core transformer, the most important component of these transistor circuits, is actually simpler than the paired pulse circuits, and is given in Fig. 2. A pulse width of 60 microseconds is needed by GC 10B and this is adjusted again by choosing a suitable value for C. The oscillograms may be seen in Fig. 4b. The crystal diode across transformer primary (Fig. 5) not only clips overshoot, but also conserves current drain, as it returns a good part of the magnetic energy back into the supply.

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X-RAY STUDY OF CRYSTALLITE ORIENTATION IN AGAVE AMERICANA

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Plate VI

ABSTRACT. Following the method outlined by P. H. Hermans and others, a quantitative study of crystallite orientation in agave americana fibre under various physical conditions is carried out. It is found that unlike many other cellulose fibres, the orientation of the crystallites in agave americana shows improvement on treatment with NaOH. This is ascribed to the removal of some intercrystalline constituents.

Because of their textile importance cellulose fibres of different kinds and origin are probably the most extensively investigated of all the natural materials. They are known to occur as Natural, Bast and Leaf fibres. The determination of crystallite orientation in a particular type of leaf fibre-agave americana forms the subject matter of the present investigation.

The problem of crystallite orientation in fibres is not only of theoretical interest but also of practical importance as many of their physical and chemical properties are correlated with it. A quantitative investigation of the problem has been attempted by Sisson and Clark (1933), Berkley (1939), R. Hosemann (1937) and Hermans and Hermans (1946). The problem has been studied both from X-ray and optical methods. The X-ray method is more useful because optical methods furnish information merely as to the average orientation of the entire fibrous substance, whereas the X-ray method gives orientation of only the crystallite components. It is in this restricted sense of orientation that a quantitative estimation by means of X-ray methods is made.

The intensity distribution along an X-ray interference circle from a polycrystalline specimen in which crystallites are oriented in some way, offers a possibility to investigate the orientation properties of the material. This was first pointed out and the foundations for such investigation were laid by Polanyi (1921). Since that time a very large number of orientation investigation has been published dealing with inorganic and organic materials of crystalline and semi-crystalline nature. Following Hermans, and Hermans, the orientation factor is determined by the equation

$$f_x = 1 - \frac{3}{2} \left(\overline{\sin^2\beta_1} + \overline{\sin^2\beta_2} \right) \qquad \qquad \dots \quad (1)$$

where β_1 and β_2 are the angular distances along the equator for two paratropic interferences arising from planes approximately at right angles to each other. In taking averages along the arcs we assign weights $G(\beta) \cos \beta_1$ and $G(\beta) \cos \beta_2$ the value of β according to the shape of the intensity distribution $I = G(\beta)$. Thus

$$\overline{\sin^2\beta} = \frac{\int I \sin^2\beta \cos \beta \, d\beta}{\int I \cos \beta \, d\beta}$$

However, since in those cases where (101). (021) overlap the intensity curve for (10$\bar{1}$) alone can not be determined. the following formula is used instead of (1)

$$f_x = 1.245 - 1.72 \sin^2\beta - 2.06 \overline{\sin^2 t}$$

where t is the angle along the (101)--(021) circle relating to the total intensity of two overlapping interferences. This is based on $K = 0.83$

EXPERIMENTAL

Raw leaves of agave americana were retted as usual and the fibres obtained where thoroughly washed. dried and combed to ensure parallelism. Filtered CuK_α radiation from a Seifert's sealed tube working at 38 KV, and 18 mA was used with a specimen to film distance of 5 cm. and the specimen size, exposure time and photographic technique were standardised as far as possible. The photometer used was Moll's recording type. The film was cut into a circular disc of 4.5 cm diameter and it was mounted on a rotatable holder fixed to the stage of the microphotometer so that the rotation which could be made in steps of 2° arc took place about the centre of the photograph. At each setting the film was scanned radially by traversing the holder. Since the films were of low photographic density it was assumed in the calculations that the X-ray intensity was proportional to blackening and was thus linearly related to the logarithm of the intensity of transmitted light. The results obtained are given in Table I.

TABLE I

Sample	$\sin^2\beta_1$	$\sin^2\beta_2$	f_x
Native Fibre	.041	.055	.85
When treated with 18%NaOH and dried without tension.	.026	.0406	.90
Treated with 18% NaOH and dried under tension	.016	.014	.96

Native fibre boiled with 2%H_2SO_4 band mercerised with 18% NaOH = 0.75

DISCUSSION

Agave americana fibres are stiff, bright and comparatively thicker than jute in the native form. On treatment with caustic soda solution the strength
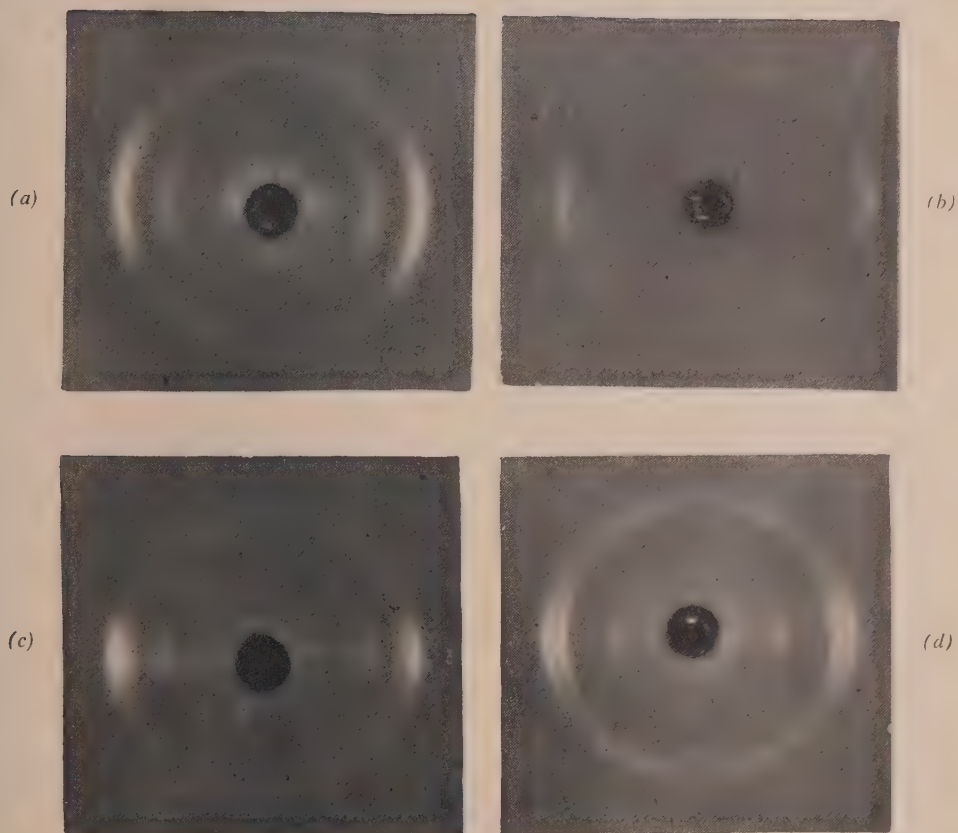
Fig. 1. X-ray photograph of the fibre

(a) Native fibre
(b) Treated with 17% NaOH, washed and dried
without tension
(c) Same as above when dried under tension
(d) Boiled with 2% H2SO4 and then
mercerised.

of the fibre gradually decreases which is evidently due to the removal of lignin and other intercrystalline materials which may also be responsible for an incomplete mercerisation. This is clearly seen from the X-ray photograph of the fibre treated with 18% NaOH (Plate VI). The photograph is a mixed one. The partial reconversion of the cellulose to the native modification can as well be ascribed to the formation of crosslinks between cellulose chains in the neighbourhood of the crystallites and thus creating a disposition in favour of the return to the original configuration after swelling. This seems to be supported by the fact that the degree of mercerisation is more pronounced in a sample which is boiled with 2% H_2SO_4 before being mercerised because it loosens the crosslinks. There is, however, also the possibility of the removal of some intercrystalline constituents which effect mercerisation. Another interesting observation made about agave americana is that the crystallites are better oriented as a result of the treatment with NaOH. This may again be due to the removal of those intercrystalline materials which impede the rotation and alignment of the crystallites. However when the fibre is first boiled with H_2SO_4 and then mercerised, the orientation decreases. This shows that the damage done on H_2SO_4 treatment far outweighs the improvement in orientation when NaOH alone is employed.

A detailed study of agave americana from the scattering of X-rays at small angles and by employing the techniques of electron microscopy is also being made and will be shortly reported.

ACKNOWLEDGMENTS

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2

SOME OBSERVATIONS ON THE ENERGY SPECTRUM OF LOW ENERGY BREMSSTRAHLUNG FROM ELECTRONS OF ENERGY $\leq 10^{12}$ eV

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ABSTRACT. Observations have been made on the energy spectrum of photons of energy greater than 30 MeV emitted by bremsstrahlung from high energy electrons of 10^{11} eV to 10^{12} eV. This observation is of importance from the point of view of checking the influence of the density of the medium. No significant departure from the conventional theory has been observed.

INTRODUCTION

In the past few years, a new aspect of the electrodynamic theory of radiation has evolved from the theoretical prediction of the Russian physicists Landau and Pomeranchuk (1953) and Ter-Mikaelyan (1954), according to whom the probability of bremsstrahlung of low energy photons depends upon the density of the medium. In the conventional theory due to Bethe and Heitler (Heitler, 1954) there is no such dependence. According to the new effect, abbreviated as L-P-T, the cross section for the emission of low energy photons is predicted to decrease in media of higher density, as higher initial energies for the primary electron are approached. This influence results from the multiple scattering of the electron and due to the polarisation of the medium. On the basis of quantum mechanical considerations, Migdal (1957) has worked out the details of the effect. Curves suitable for comparison with experiment have been given by Varfolomeev *et al.* (1958, 1959)*

Nuclear emulsions have a density of ~ 4 g.cm^{-3} and electromagnetic cascades initiated by isolated electrons or photons and by the photons from the π°-mesons created in very high energy interactions are readily available. For primary electron energies greater than or $\sim 10^{11}$ eV and secondary electron pairs due to photons of a few MeV, one expects to check the validity of the theoretical predictions. This article is meant to describe the results of our work and discuss these in the light of the other results available on the subject.

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* Thanks are due to Professor Varfolomeev for the communication of the preprint and the reprint.

STATEMENT OF THE PROBLEM

It is important to consider the problem in the light of the experimental observations that can be made. One normally observes a soft shower and obtains its energy from parameters that involve the growth of the shower which is derived from the longitudinal development as given by the cascade theory or from the lateral spread (Pinkau, 1956). There are also alternative methods such as the suppression of ionisation near pair origin (Iwadare 1958) or the true opening angle of the pair (Lohrmann 1955; Aditya 1959 a). The second problem is to detect the secondary pairs and determine the energy of each one of them. When both these quantities are known, the theory can in principle be put to test.

It is well known that the intrinsic fluctuations (Aditya 1959 b) involved in the nature of the processes do not always permit a precise estimation of the primary energy. Results derived from one or a smaller number of cascades are thus subject to uncertainties. This limitation can be overcome to a great extent by collecting together a large number of cascades of about the same energy. Another factor that plays a decisive role, is the probability for the detection of low energy electron pairs. In nuclear emulsions the critical energy is ~ 20 MeV so that one is not likely to detect pairs of energy smaller than this value with as good an efficiency as the high energy pairs. The detection efficiency is strongly dependent upon the experimental conditions of observation.

There is yet another factor that influences our conclusions on the effect, as follows : The lack of low energy pairs is strongly dependent on the primary energy, so that one would like to take account of only the first generation pairs due to the bremsstrahlung from the primary electron and not those created from the secondary electrons. This distinction between the pairs of the various generations is not straightforward, but has been attempted by some workers (Bonisz *et al.*, 1959; Fenyves *et al.*, 1959). In such a procedure there is a possibility of introducing a bias towards the removal of more low energy pairs than the high energy one's. In the light of the considerations given above, the results of the present investigation are given.

EXPERIMENTAL DETAILS

Out of a large number of soft showers picked out from two stacks of stripped emulsions exposed in the stratosphere (Aditya 1959 c), 17 cascades have been selected, the criteria for selection being good conditions of experimental observation. There is apparently no bias likely to affect the conclusions on the subject. 3 cascades are associated with a nuclear disintegration (Aditya 1959 d), 5 are initiated presumably by a single electron entering the stack and 9 originate from a single pair. In each case the development has been normalised to a primary electron and the cascades grouped into two bunches : high energy (650, 625, 550, 500, 500, 475, 350, 325)and low energy (250, 225, 200, 125, 50, 50, 40, 40, 40) groups.

The figures in the brackets give for each cascade the energy per electron in GeV. In the case of cascades initiated by a pair, the energy has been assumed to have been equipartitioned between the two electrons. These energies have in most of the cases been determined by the application of different procedures and the most probable value estimated. The individual errors in the energy estimation are expected to have been smoothed out as the cascades of about the same energy have been grouped together.

In order to keep the detection efficiency presumably constant over the entire energy region, we have not attempted to consider the electron pairs of energy less than 30 MeV. The resulting energy spectrum up to a distance of 1.5 cascade unit has been plotted in the Figs. 1a and 1b, for the two energy intervals. The expected curves for the Bethe-Heitler and Migdal calculations have been included for comparison. We have made no attempt to separate the pairs of various generations on account of the reasons already mentioned. In order to decrease the influence of the pairs of further generations, the measurements have been limited to only the first 1.5 cascade unit from the origin.
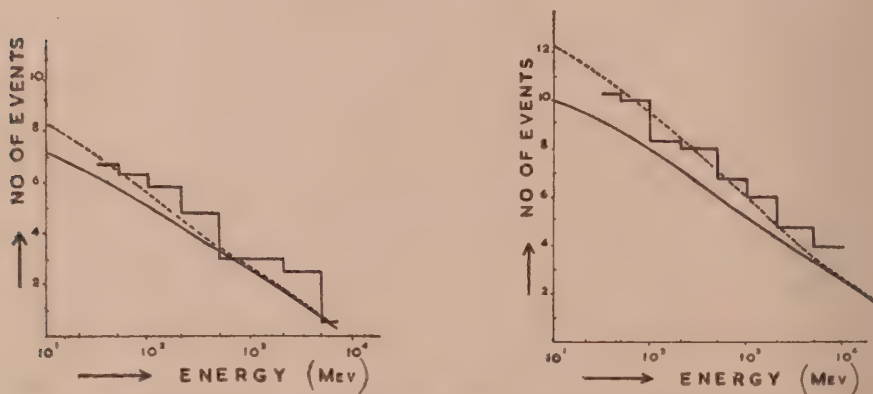

Fig. 1. Energy spectrum for electron pairs observed over the first 1.5 cascade length. The lower limit for acceptable pair energy has been set at 30 MeV (see text).

(a) 9 cascades of median energy 50 GeV per electron.

(b) 8 cascades of median energy 500 GeV per electron.

From the figure, it may be seen that subject to the experimental fluctuation a conclusive statement might not be made in favour or against one or the other theory. Since our energies are not extremely high, allowance has to be made for the possibility of the suppression being genuine at higher energies, there being no measurable departure up to ∼ 500 GeV energy for primary electrons.

DISCUSSION

It appears useful to compare our results with those of other workers in the field. Varfolomeev *et al.*, (1958, 1959), have studied the spectra for primary

energies from 10^{11} eV to 10^{13} eV, and secondary pair energies up to 1.5 MeV. They have found a significant departure from the Bethe-Heitler theory and agreement with L-P-T. It is worth considering that their lower limit of ~ 1 MeV is too low to guarantee uniform detection efficiency over the entire range. In view of this it may not be fair to consider their measurements as having established the existence of the effect.

The second investigation is that of the Polish group (Benisz *et al.*, 1959) who have studied 4 photon-initiated cascades out of which three are associated with a high energy disintegration and the fourth one is that of Miesowicz *et al.*, (1957). The mean energy per electron is ~ 500 GeV, and they have attempted at the separation of the pairs of the first generation from the rest. The lack of low energy pairs might well be accounted for the introduced bias. Their energy region is the same as ours and since our statistics is relatively larger with no effect observed, it may be concluded that the effect if present at these energies is not at least of the order suggested by L-P-T.

There is yet another work by the Czech-Hungary group (Fenyves *et al.*, 1959) in which they have studied the energy spectrum up to 1.5 cascade unit for a cascade initiated by a photon of ~ 2000 GeV. Inspite of the fact that this energy is fairly high (in fact higher than our energies and of Benisz *et al.*) no departure has been noted. The authors have attempted at the separation of the various generation pairs and still observed no divergence from the conventional theory.

In view of the present investigation and of the investigation of other workers mentioned above, it may be concluded that the decrease of bremsstrahlung cross section for low energy photons is not appreciable. This work however does not prove whether the departure would exist at higher energies.

After this work had been finished, the results of the Bristol group (Fowler *et al.*, 1959) have come to our notice*. The method is based upon measuring the average distance of materialisation of the first pair for two groups of cascades of different energies. From the distribution of these distances and the mean value for two groups of primary energy 10^{11} eV and 10^{12} eV, they found better agreement with L-P-T than with B-H. Their energy of 10^{12} eV per electron is much higher than most of the other investigations and in view of the large statistics give evidence on the existence of the effect at $\sim 10^{12}$ eV. The actual magnitude of the suppression, whether it is as much as predicted, will have to be determined from the availability of larger statistics.

P. K. Aditya

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A HIGH INTENSITY NEUTRON GENERATOR

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ABSTRACT. A 400 KV deuteron D.C. accelerator of Cockroft-Walton type, for production of neutrons by $d-d$ and $d-t$ reactions, is described. Special features and principal operating characteristics of this generator are discussed.

I. INTRODUCTION

A 400 KV Cockroft-Walton type deuteron accelerator, designed for delivering 5 mA of deuteron ion beam, had been planned, fabricated and installed in this institute and been operating regularly and efficiently for the last eighteen months. The exothermic nature of (d, d) and (d, t) reaction and its high cross section made this type of generator compact and low voltage one. The development and easy availability of the targets of deuterium and tritium gas absorbed in Zirconium or Titanium, have given a strong impetus for building this compact low voltage unit for producing fairly strong sources of monoenergetic γ-free neutrons in the 3.5 MeV and 14 MeV range. In fact, several reports (Peek and Eubank, 1955; Bergstralh, *et al.*, 1953; Lorrain, *et al.*, 1957; Bonner, *et al.*, 1959) of such generators have appeared in recent years in the literature. In our case, the aim was to produce and accelerate the most intense beam of deuteron ion that can be brought to bear on the target of absorbed gas quite safely. To achieve this end, we have developed the ion source and the lens system in such a way as to focuss an unanalysed deuteron beam of 2.5 mA. Also the cooling of the target was thoroughly investigated so that in the end we could focuss on the target a beam current of more than 1 mA. The principal characteristics of our generators are (i) its high ion beam consisting of more than 90% of monatomic ions, obtained by suitable modifications of ion source. (*ii*) The high voltage is obtained by multiplying a high frequency voltage in a cascade column. This is done both for the purpose of economy and reduction in size as also for reduction in ripples and load effect. The filament supply of the rectifier column is separate and also of high frequency. (iii) The cooling of the absorbed gas target is directly by refrigerant or liquid nitrogen, with an eye to the maximum possible heat transfer, so as to bear as high an ion beam as possible on the target without exceeding its safe temperature of 70° at any time even locally.

II. C–W VOLTAGE GENERATOR

The voltage generator is, as usual, a voltage multiplying circuit based on the original Greinacher (1921) circuit, as improved progressively by Cockroft-Walton (1932), Gradstein (1936), Arnold (1950), Lorrain (1949), etc. Particularly the masterly analysis of voltage multiplying circuits, by Bowers (1939), and by Mitchell (1945) have contributed handsomely towards the understanding and improvements of this type of voltage generator. Following the development by Douma and Brekoo at the Philips laboratory and the suggestions made by Lorrain, radio frequency was adopted for both the main High Voltage and also for heating the filaments. We discarded the practice of using power frequencies or other near frequencies. In addition to the marked economy in cost and equipments, a considerable reduction in size was achieved by the use of radio-frequencies. Also a decided improvement in the matters of ripples on the output voltage and of voltage drop with load, was achieved through its use. Although the presence of voltage ripple is not a serious handicap to its use as a neutron generator, still the smallest value of ripple voltage is an advantage in achieving a sharply defined energy. In fact, the higher the frequency and the larger the capacity, the smaller will be the ripple and load effect.

The output voltage of a c-w multiplier unit can be written as

$$V = 2nE - \Delta V - \delta V.$$

$$= 2nE - \frac{i}{fc} \left(\frac{2}{3} n^3 + \frac{1}{2} n^2 - \frac{1}{6} n \right) - \frac{i}{fc} \left[\frac{n(n+1)}{2} \right]$$

$$= 2nE - \frac{i}{fc} \left[\frac{2}{3} n^3 + n^2 - \frac{n}{6} + \frac{1}{2} \right]$$

$$= 2nE - \frac{i}{fc} \cdot \frac{n}{3} (2n+1)(n+1) \qquad \qquad \dots \ (1).$$

where n = number of stages in the multiplier column
E = peak input voltage
ΔV = voltage drop due to load
δV = ripple voltage
i = current drain on the high voltage
f = operating frequency
C = condenser value

The effect of higher condenser and frequency values is apparent. So also the desirability of limiting the n value. In practice, there is an upper limit to the frequency that can be usefully employed. Also the value of the condenser is decided by the bulk and the cost. Some compromise is made between these conflicting factors and it was found that 100 Kc/s and 400 Kc/s are two of the best practical values for the mains and filament heating frequencies. The theo-

retical values of ripple voltage is $3.5V$ per mA of load current, and of voltage drop
due to load is 22 volt per mA, assuming the value of $n = 7$ and $c = .05\mu f$.

The unit of a multiplying circuit of our generator is shown as in the Fig. 1.
The C-W generator has several pairs of Eimac 100 R radiation cooled rectifier
arranged in cascade supported by equal numbers of condensers arranged in
column. The tubes are encased in leaklight perspex tubes coupled to each other
by aluminium domes. The other arrangements of the column are not far different
from that adopted at Philips laboratory. The filaments of the tubes are heated
by 350 Kc/s power coupled by ferroxcube core transformer. Plate power supply
frequency is 75 Kc/s. The isolation of the two frequencies from each other was
obtained by means of LC circuits resonant at the filament heating frequency.
The multiplier circuit is supplied by a master oscillator and power amplifier—
the output of which is amplified by a very high Q resonant coil, which in turn
feeds the multiplier column. The bleeder resistance is a chain of five resistances
of 100 meg ohms each, which draws a current of 800 μA at maximum voltage of
400 KV. This column of resistances is also encased in a perspex tube to be filled
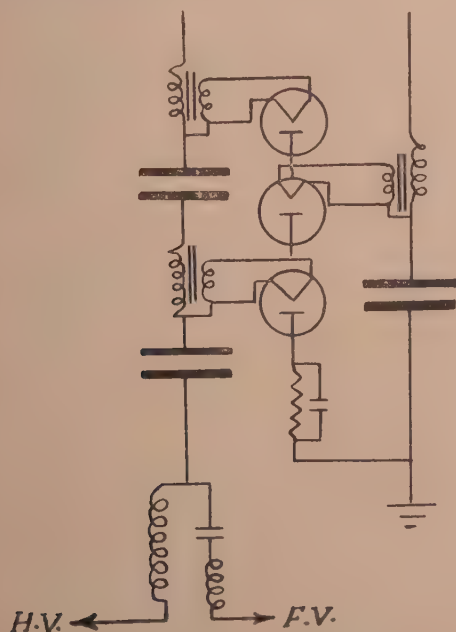with silicone oil for better stability.

Fig. 1.

The total resistance of the column was measured within an error of 0.1%.
It was observed that the resistance column value remained fairly constant over
a long period of time. Percentage of ripple was determined by means of an

3

oscilloscope, taking the output from the low voltage end of the resistance column. The measured value was found to be 7 volt per mA of load current. The voltage drop with 7 mA of load current is 140V. This agrees within close limits with our theoretical value as calculated from the formulae shown in Eq. 1. The stabilisation of the high voltage was achieved by the stabilisation of the input R.F. voltage from the oscillator. This in turn was done by taking a small fraction of the voltage output and using them to correct any change in the D.C. plate voltage of the oscillator. The power to the filaments of the transmitter tubes was fed through a magnetic voltage stabiliser. The actual stabilisation achieved in this way was better than 0.1%.

GENERAL ARRANGEMENTS

The plan of the general arrangements of the high tension set, the accelerating unit and the power supplying alternator is shown in the Fig. 2. The accelerator column consists of three numbers of silica tubes. Detailed description of the accelerating tube is given in the following section. The accelerating column
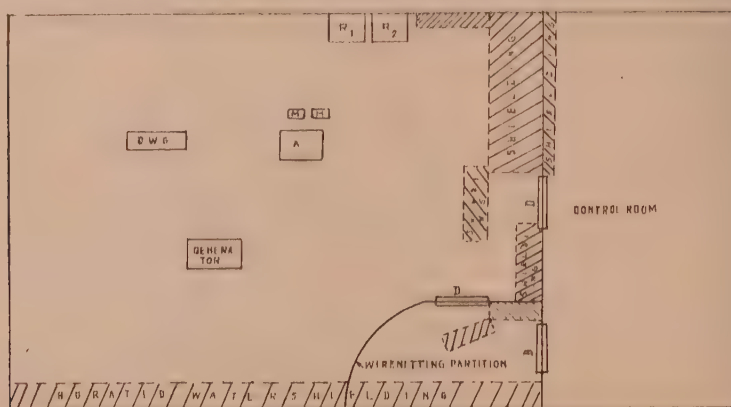

Fig. 2.   General plan of layout of C-W Generator.

is placed vertically in our case — though we considered the advantages accruing from the horizontal alignment of the column which is very often followed in the case of low voltage accelerator. We are guided in this respect by the ease of assembly and alignment in the vertical system as also the ease of automatically providing high voltage insulation from the ground. The units for supplying power to the ion source and the focussing lens—all should be at high voltage and hence placed in a housing well insulated from ground. The high voltage terminal housing is a polished aluminium dome rounded at corners to prevent corona loss. The four insulating columns for supporting this high voltage terminal are all made of 3″ dia. perspex tube sealed at both ends to prevent penetration of dust and moisture. The outside surface of these tubes are treated and polished

with silicone fluid D. C. 200 to prevent any adhesion of moisture on them. Inside the high voltage terminal are located (a) the radio-frequency ion source oscillator and its power supply, (b) the extracting voltage supply (0—5 KV), (c) the focussing voltage supply (0-15 KV) and (d) the cooling fans and light arrangements. The precision leak valve and its control together with the gas reservoir are also located inside this dome. Appropriate meters to indicate operation of the above circuits are suitably placed in the dome with clear perspex windows. Control rods in the form of perspex tubes come out of the housing from each circuit. The tubes are operated by remotely controlled system. Mirror and telescope arrangements were made to obtain remote view of the H.V. terminal operation. An alternator for supplying power to the units in the high voltage terminal, is placed in a dome similar to the above. The alternator generates 110 V. A. C. single phase, 800 cycles frequency. The alternator body is made of aluminium alloy, thereby ensuring a light weight. The alternator rotates at 3000 r.p.m. and is driven by a motor placed at ground end and the driving system requires careful designs and high insulations.

In our case this is driven by a 3 H.P. A.C. 3 phase motor with speed of 3000 rpm. The coupling system is a $2\frac{1}{2}''$ dia. perspex tube, 4 ft. long, suitably joined at the ends by means of universal couplings. No speed reduction gears are employed now. This arrangement is marked by simplicity and directness, though it required very careful adjustments at the beginning. An alternate arrangement involving the reduction of speed of rotation of perspex shaft by a factor of 10, is in progress.

ION SOURCE

Several types of ion sources were examined for suitability of their uses in this particular instrument. Of these, two types immediately recommended themselves by reasons of stability, long life and ease of maintenance. One of them is the Radio-frequency ion source and the other the Penning Cold Cathode ion gauge type. For low power consumption, circuit simplicity and compactness, the penning type source seemed better. But for high ion yield, and for high percentage of atomic ion in the beam, R. F. type source has hardly any equal. Since our instrument requirement is for highest atomic ion beam consistent with low gas consumption, R. F. ion source became our obvious choice.

In our design, we adopted the Oak Ridge type with some modifications for higher ion yield. The discharge tube is a $1''$ dia. Pyrex glass tube, $6''$ long, sealed to the metal plate by means of gasketed flanged joints. The Extraction canal is of pure aluminium $1/16''$ dia. $\times 3/16''$ long. The cover glass for the canal is of pyrex, —its top grounded flat and polished (Fig. 4). A triode 3/300 tube was used in an ultra-audion circuit to provide a maximum power output of 300 watts at 75 mc/s. The coupling was done by quater wave transmission lines ; glass

leak to the tube was controlled by a finely adjustable leak valve. A strong magnetic field was provided around the canal tip. The life of the glass tip and the aluminium canal depends greatly on the beam current extracted throughout it. In our case the averge life of the canal and the tip is more than 200 hrs. The part played by cleaning of the discharge tube can not be over emphasised. The method of cleaning by diluted hydrofluoric acid was found to be good and occasional cleaning by this method tended to increase the ion output.

Deuteron currents approaching 4.5 mA at 4 KV extraction potential could be obtained from this ion source. The beam divergence was small enough to be focussed by an electro static lens within a spot of 3-4 mm at a distance of 1 meter. Of course this required increased gas feed to the source and a longer canal diameter. Experiments on monatomic ion percentage in the deuteron beam current showed that quality of glass of the ion source and its annealing conditions have a strong influence on the high monatomic ion percentage. It was also found that after more than 200 hours running, the ion source tended to produce a lesser yield of atomic ion percentage. This is probably due to vitrification of the glass material.

VACUUM SYSTEM

The pumping unit initially consisted of one 4″ oil diffusion pump backed by a rotary mechanical pump having a free air displacement capacity of 140 lit/min. The oil diffusion pump was designed and fabricated by us and it developed an unbaffled pumping speed of 275 lit/sec at 10^{-5} mm Hg. The mechanical pump developed a backing pressure of 6×10^{-4} mm Hg and speed of 0.7 lit/sec when the system was running with no gas load. A refrigerated optical baffle using Freon-22, was employed to trap all the back streaming oil vapour and keep the accelerating tubes and target chamber free from traces of oil vapour. The actual pumping speed developed with this baffle and system was 140 l/sec at 1×10^{-5} mm Hg. To handle increased gas feed to the ion source at the time of increasing ion beam current, and also to handle occasional gas bursts, provision of another 4″ diffusion pump was made. This later pump was designed with an eye to its ability to handle larger throughput. The baffle system of this pump was made in two stage. One was a water baffle and the second a freon cooled refirgerated baffle. The temperature of the water baffle was designed to be of 35°C.

The vacuum measuring gauges consist of one thermo-couple gauge for low vaccum range (.1 mm to 10^{-3} mm) and one hot cathode ionisation gauge for high vacuum range (10^{-4} to 10^{-6} mm Hg). Adequate provisions of ion gauge control, interlock system of high vacuum with ion gauge, water flow, refrigerating cooling unit and diffusion pump heater, were arranged. Weston sensitrol relays were freely used for these control. The r.f. ion source at its normal operation consumes nearly 10-15 cc. of gas at atmospheric pressure per hour. This means a leak

load on the high vacuum side of the accelerator tube. The ultimate vacuum in the accelerator tube with no gas load, as measured by the ionisation gauge at the ground end of the tube, was 2×10^{-6} mm Hg and the vacuum with the ion source and the system running, was better than 1×10^{-5} mm Hg.

ACCELERATING COLUMN

The accelerating column forms the most important item in the neutron generator. This consists of three silica tubes each of 6″ i/d × 10″ long with wall thickness of 1/4″ and flanged at each end. These are joined together by means of aluminium plates having grooved gasket on each face. The plates and the silica tubes are clamped together and covered by means of ring shaped polished aluminium domes. Each aluminium plate carries a lens. These lenses are formed of spun copper tube, heavily chromium plated. The shape, gap width, diameter and other configuration of the lenses are given in the Fig. 3. Each gap is screened from the silica tubes wall to avoid any accumulation of electrostatic charge by

Fig. 3. Ion acceleration column.

the insulator wall. Because of the particular mechanical set up of the lense system, good alignment is automatically assured at the time of assembly. Fig. 4 is a

sketch of the focussing electrode together with the ion source assembly.   The ion source is mounted on a gasket in a plate on the other side of which the focussing system is integrally mounted by means of a high voltage ceramic seal.   The ion
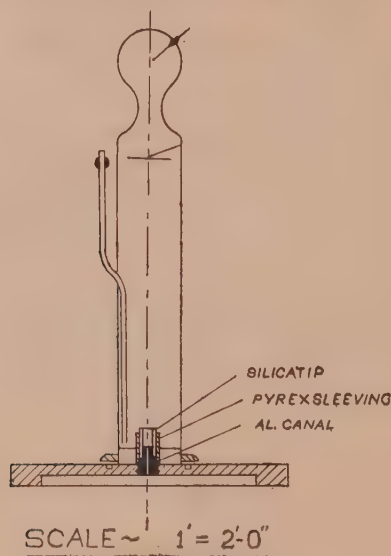

SCALE ~ ¹ 1′= 2′-0″

Fig. 4. R. F. ion source.

source can be aligned with respect to the probe canal and the focussing lens by means of a set of removable adjusting screws.   These adjustments are to be made when the system is running.   The arrangements of the lens system is such that the major part of the accelerating voltage is developed across the last lens gap. As a result the beam is focussed on the target, at a distance of 1.8 metres from the ion source, under all values of high voltage exceeding a certain minimum value. The focal length of the last lens is almost constant irrespective of the voltage on it as it falls on the asymptotic region of the f-v curve (Fig. 5).

Fig. 5. Deuteron energy in KV.

TARGET ASSEMBLY

As this instrument is a high current one and meant for high neutron flux, the design of the target system, particularly its cooling, calls for careful attention and examination of details. Our instrument is designed to operate with either tritium or deuterium target. For (D, T) reaction, Tritium gas absorbed in titanium and mounted by evaporation on copper, was used. Both the thick and the thin types were employed, depending on the necessity of particular experiments. The useful life of a target depends on the efficiency of the cooling system and its ability to transfer the maximum amount of heat without allowing the target to exceed the safe temperature of 70°C even momentarily. The development of hot spot on the target is to be carefully avoided.

We have used several types of target mountings and cooling arrangements. First one we used is shown in Fig. 6(i). The cooling was done by precooled water forced directly on the underside of the target in a jet at a maximum pressure of 30 lbs/sq. in. The target intercepts the beam at an angle of 45°, and also there is an arrangement to rotate the target at a slow rate by means of motor and gear assembly. An offset in the target manifold enabled one to bring the sample for bombardment within 4 cm from the target. The performance of the target is such that at 200 Kv up to 450 μA of unanalysed deuteron beam can be brought to bear on the target. The neutron yield under this condition is about 4×10^{10} n/sec. The target temperature rose up to 70°C. The mechanical pump output was monitored for traces of degassed tritium. The target life was understandably short under this condition.
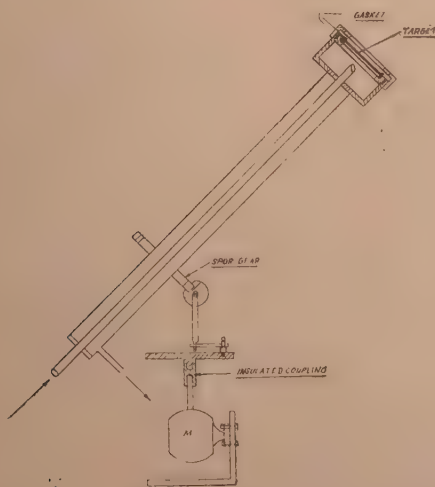

Fig. 6(i). Water-cooled oscillating target.

We changed this target arrangements in favour of another in which cooling is done by means of a refrigerant. The arrangement is shown in Fig. 6(ii). The minimum temperature that the target attains is 8°F. The gas used is Freon 20, and the capacity of the refrigerating unit is more than 4000 BTU hr.; under this condition more than 850μA of deuteron beam at 200 KV, was put on the target and continuously operated. For short run not exceeding 1 hr. at a time more
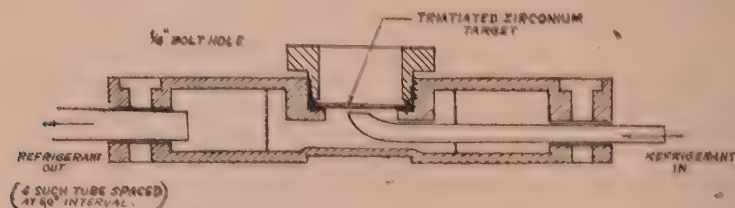

Fig, 6 (ii).   Target cooling system. Refrigerant-cooled target.

than 1.5 mA was tolerated without exceeding the safe limit of target temperature of 75°C. Temperature measurement was done by means of a pair of differential thermocouple attached to the target. The maximum neutron yield obtained was more than 3×10^{11} n/sec. The beam on this target which is not a rotating type, is purposely defocussed on an area of approximately 3 4", and is thus designed to cover the entire useful area of the target. In this target the bombarding sample can be brought within less than one cm of the target.

Still another type of target cooling was designed for use with liquid nitrogen coolant, but this was discarded in favour of the refrigerant cooled unit in view of the frequent attention that the former type requires. In all these types the main source of trouble is the deposition of carbon, the target assembly due to the traces of oil vapour present from the diffusion pump. This trouble was eliminated in the refrigerant cooled unit by providing a cooler guard ring around the target on which most of the vapour deposit, thus keeping the target free from contamination.

ADJUSTMENT

The ion acceleration system needs very careful adjustments and precise alignment in order to obtain the maximum ion beam on the target. This is important, otherwise several undesirable characteristics will appear. Some of the more important of such unwanted characteristics are :

(i) the radial and axial drift of the focal point of the ion beam, with change in acceleration voltage,

(ii) change or fluctuations of accelerating voltage with variation in load.

These factors were eliminated with proper care in design and alignment. The ion acceleration systems were first mechanically aligned to a high degree of

precision with respect to the last lens. This was supplemented by a further adjustment in the running condition. The adjustment was made to make the ion beam maximum and by this method the ion optical axes of the ion source, focussing lens and extraction probe lens were brought in one line.

MEASUREMENT OF ION CURRENT

Accurate measurement of ion beam current free from all ambiguities due to secondary electron emission and additional secondary ion collection, is very important. Due care and precautions were exercised to measure this. The most direct way is to use a Faraday cage and this cage was used in preliminary stages to measure the current. When ion beam current is high, the colorimetric method is convenient. We have measured the rise in temperature at target, by means of a differential thermocouple. The ion beam current could only be held steady to within 0.5 to 1% under good conditions. The ion current was found to be very sensitive to any change in pressure in the ion acceleration tube and steady beam current could only be obtained if occasional gas bursts in the accelerator tube is kept to a minimum. To this end, twofold means were employed :

(i) keeping the vacuum system scrupulously clean and chromium plating the lens system,

and (ii) using a diffusion pump unit of high enough speed, capable of handling a large thoroughput without undue rise in ambient pressure.

NEUTRON FLUX AND SPECTRUM

The neutron flux was measured by three methods :

(i) by means of a calibrated neutron counter

(ii) by means of threshold detectors

and (iii) by means of nuclear emulsion plates.

In the first method. we used a BF_3 counter (containing enriched B^{10}) in a long counter geometry. This counter was previously calibrated very carefully. Its advantage is that it is intensitive to γ-ray and inherently stable and has very low background, and disadvantages are that it is sensitive to neutrons of lower energy and hence could not discriminate against scattered neutron background.

In most of our experiments. the fast neutron flux measurements were made by activation of some selected element. based on neutron threshold reaction. Its main advantages are (i) it is insensitive to neutrons of energy below the threshold. (hence to most of the scattered neutrons) and (ii) it is capable of measuring flux without causing significant perturbation in flux distribution. In our case 9.9 min activity of Cu^{63} (n, 2n) Cu^{62} was used for this flux measurement. The threshold of this reaction is 10.65 MeV and the adopted value of its cross-section

4

is taken to determine any 5.1 min activity of $Cu^{65}(n, \gamma)Cu^{66}$ if it was produced, since the presence of low energy neutron or thermal neutron may cause this later activity to contribute to the total activity and cause errors. However, since Cu^{62} is positron emitter the separation of this activity from that of Cu^{66} is not
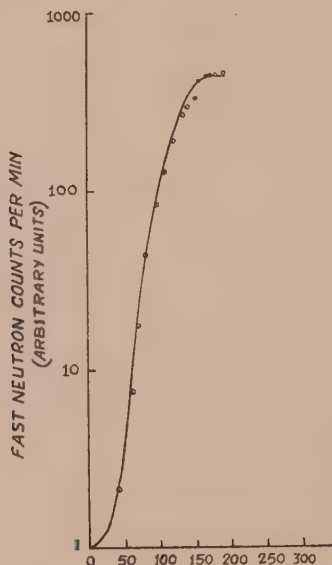

Fig. 7.   Neutron yield curve as function of deuteron energy

difficult. Where bombardment time is longer (i.e. more than 30 min) we have used the 14.8 hr. activity of Al^{27} (n, α) Na^{24} having an effective energy of 8.1 MeV for calibration. Na^{24} being both β and γ active, we have measured the β-activity by means of an end window G.M. counter and the γ activity by means of a γ-spectrometer. The results from the two measurements agree closely.

The nuclear emulsion plates were used to measure the neutron fluxes as well as the spectrum of neutrons emerging from $D-T$ reactions. We used Ilford C_2 plate of emulsion thickness of 200μ and these are exposed at an angle of $5°$ to the incident neutron. The measurement of the proton tracks released by the incoming neutrons in the emulsion, yielded the fast neutron spectra [Figs. 8(i) & 8(ii)] with plate both shielded and unshielded are given in the figure. The spectra from an old target is also given. The presence of $D-D$ neutrons (of 3.5 MeV energy) can be seen.

The degree of reproducibility and accuracy in flux determination by the above methods, have been checked by several independent measurements. A typical determination of the flux by means of several methods is given in Table I.
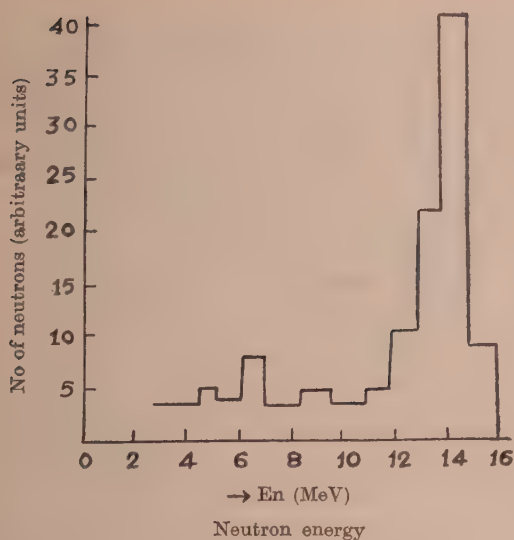
Fig. 8. (i)   Spectrum of neutrons from H³ (d,n, He⁴ detector in a channel of water and paraffin shield).
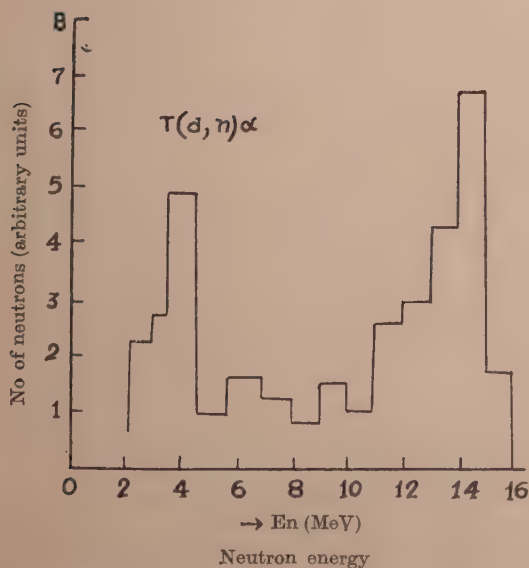

Fig. 8(ii).   Spectrum of D–T neutron from an old target , Detector unshielded.

TABLE I

Operational condition	Method of measurement	Value of neutron flux in n/sec.
Deuteron beam Energy = 190 KeV	irradiated Cu foil and measurement of β^+ activity of Cu^{62} from $Cu^{63}(n, 2n)Cu^{26}$ reaction	8.7×10^{10}
Deuteron beam Current = 580 $\mu\bar{a}$	irradiated Al foil and measurement of β^- activity of Na^{24} from $Al^{27}(n, \alpha)Na^{24}$ reaction	8.5×10^{10}
	irradiated Al foil and measurement of 1.38 MeV γ—activity of Na^{24} from the above reaction	8.3×10^{10}
	Nuclear emulsion plate Ilford $C2$ 200μ thick	8.5×10^{10}
	Calibrated BF_3 long counter	9.2×10^{10}
	Calibrated fast neutron counter	8.9×10^{10}

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INFRARED SPECTRA OF URANYL PHOSPHATE, OXALATE, AND SALICYLATE IN THE SOLID STATE

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ABSTRACT. The infrared spectra of three compounds, uranyl phosphate, uranyl oxalate, and uranyl salicylate have been studied in the region $2-24\mu$ using potassium bromide disc and nujol mull methods. About 35 bands in the case of phosphate, 25 in the case of oxalate and 40 in the case of salicylate are obtained from both these methods. The vibrational frequencies obtained from the infra-red work are correlated with those obtained form flourescence experiments.

INTRODUCTION

Corn and Wu (1938) are the first to study the infra-red and Raman spectra of uranyl acetate, nitrate, chloride and sulphate and establish the frequencies 860, 210, and 930 cm^{-1} characterising the uranyl ion. The infra-red spectra of a number of simple and double uranyl salts were studied later by Lecomte and Freymann (1941) who confirmed the above frequencies. Sevchenko and Stepanov (1949) studied the spectra of the uranyl acetate, nitrate, sulphate and potassium sulphate in the region of overtones of the above frequencies and came to the conclusion that the uranyl ion is linear in the case of acetate and nitrate and bent in the case of sulphate and potassium sulphate. All the above workers confined their investigations to the NaCl prism region only i.e., up to 15 μ.

In the present investigation, the infra-red spectra of three compounds, uranyl phosphate ($HUO_2PO_4 . 4H_2O$), uranyl oxalate [$UO_2(COOH)_2 . 3H_2O$] and uranyl salicylate, $UO_2[C_6H_4(OH)COO]_2$ have been studied in the region $2-24$ μ. The phosphate has been studied for the first time. The oxalate and salicylate were also studied by Lecomte and Freymann who, however, obtained bands, only in the region 800-1600 cm^{-1}. Even in this region, the positions of the absorption bands have been indicated in a table but the actual values of the frequencies were not given.

EXPERIMENTAL TECHNIQUE

The infra red spectra are recorded using two different methods : (1) potassium bromide disc method and (2) nujol mull method. In the case of KBr disc method

(Ford and Wilkinson, 1954), a Hilger automatic double beam prism spectrometer has been used with a rock salt prism for the region 2-15 μ. The wavelength scale is linearly calibrated in microns and the dispersion is 4.5 cm per micron. Calibration marks are made on the recorder chart, at regular intervals by a small and rapid deflection of the pen. A mixture of between 0.1 and 1.0 per cent of the sample and chemically pure potassium bromide is grinded into a fine powder and then pressed under vacuum to produce clear and transparent disc. The advantage of this method is a better distribution of very small particles in the suspending medium and the elimination of the obscuring bands which occur with mulling agents.

In the case of nujol mull method, spectra are recorded on a single beam Perkin Elmer infrared spectrometer (Model 122) with substances suspended in the nujol mull, in the region $2-24\mu$ using the sodium chloride and potassium bromide prisms. To obtain the absorption only due to substance, the transmission curve of the nujol is also recorded on the same record chart with the same experimental conditions. After substracting the absorption due to nujol, the transmission coefficients have been calculated for each point and a graph is drawn between the percentage transmission and wavenumbers.
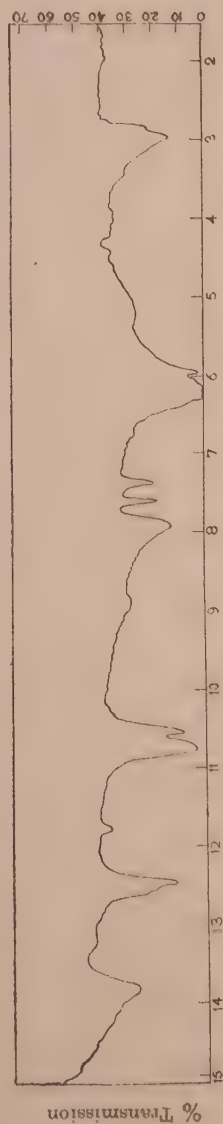
RESULTS

A comparison of the frequencies of bands obtained from the potassium bromide disc and nujol mull methods, shows that there is reasonable agreement between the two data. However, the absorption bands with the potassium bromide disc method are sharper and better resolved than in the case of nujol mull method.

The frequency values of the bands are given in Tables I, II and III and the curves are given in Figs. (1 to 9). The UO_2^{++} ion fundamentals are recorded in all cases. A few bands could be also combination bands. The large number of other bands may consist of combinations of v_1, v_2 and v_3 of UO_2^{++} ion or fundamentals, combinations and overtones of other radicals of the salts. Prominent examples are the bands due to water of hydration in the 3400-3500 cm^{-1} region and the C $=$ 0 frequencies in oxalate and salicylate occurring in the region 1600-1700 cm^{-1}.

The fluorescence and absorption spectra of these compounds are also studied by the author at liquid air temperature with powder samples (Details of these results will be published elsewhere). It is found that the phosphate and oxalate are fluorescent and the salicylate has not shown any fluorescence at all. The ground state frequencies obtained from the analyses of the fluorescence spectra are given in the third column of Table I, II, and III for correlation of the results. It is seen that there is good agreement between the various data.

Fig. 1. Infrared absorption spectrum of uranyl phosphate (KBr disc method)

Fig. 2. Infrared absorption spectrum of uranyl oxalate (KBr disc method)
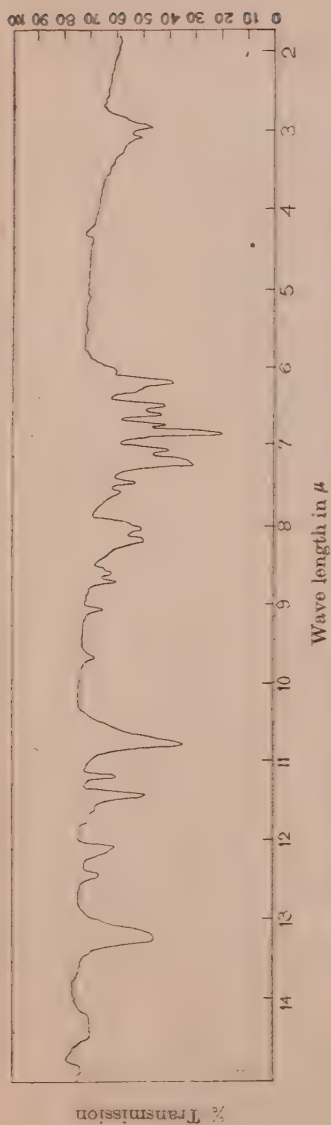
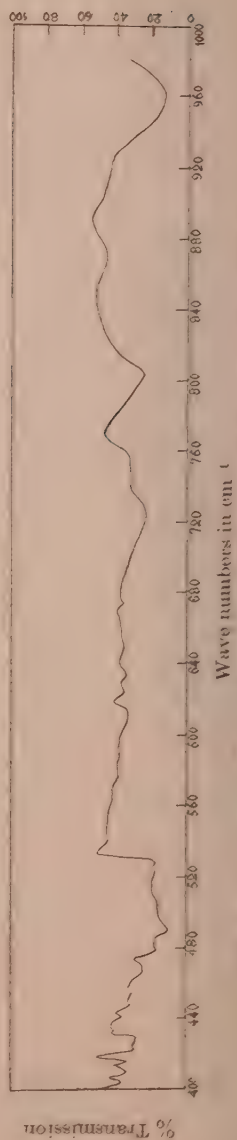Fig. 3. Infrared absorption spectrum of uranyl salicylate (KBr disc method.)

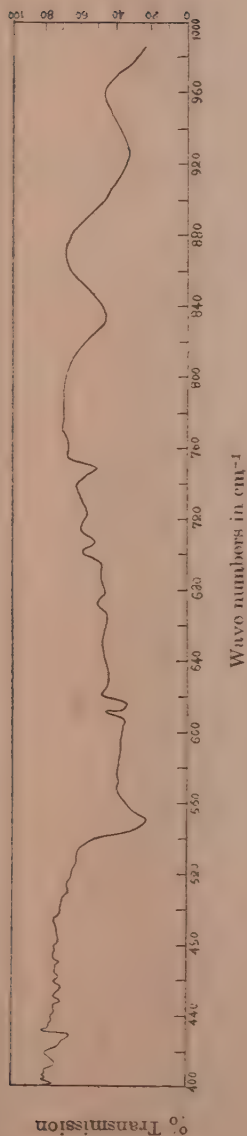Fig. 4. Infrared absorption spectrum of uranyl phosphate (Nujol mull method.)

Fig. 5.   Infra-red absorption spectrum of uranyl oxalate (Nujol mull method.)
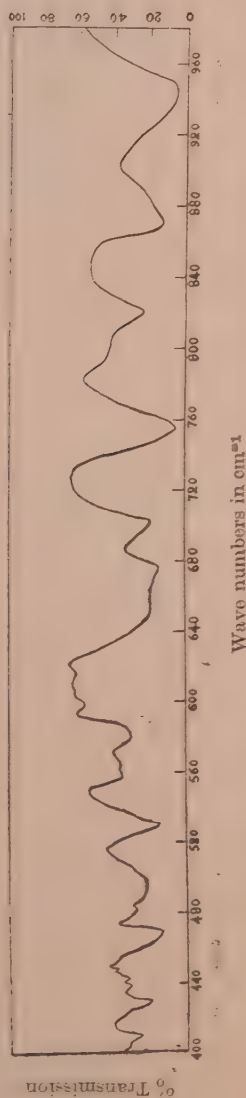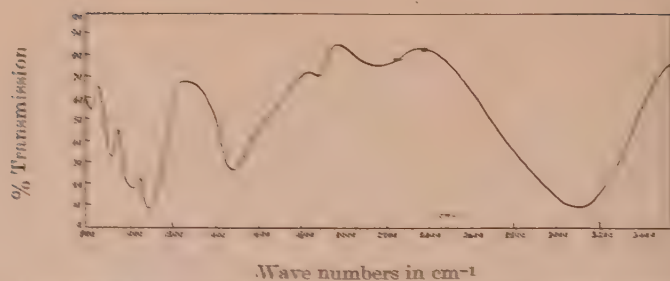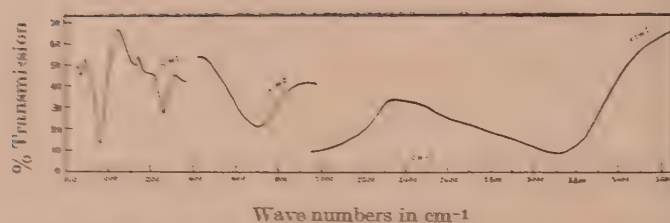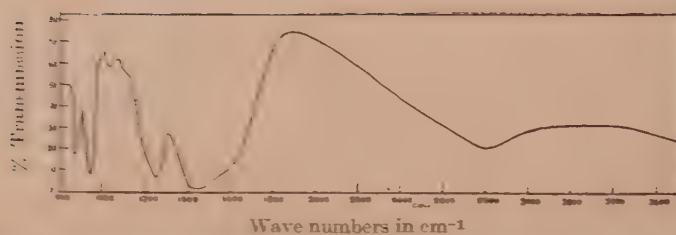
Wave numbers in cm^{-1}

% Transmission

Fig. 6.   Infrared absorption spectrum of uranyl salicylate (Nujol mull method)

Wave numbers in cm^{-1}

% Transmission

5

Wave numbers in cm⁻¹

Fig. 7. Infrared absorption spectrum of uranyl phosphate (Nujol mull method)

Wave numbers in cm⁻¹

Fig. 8. Infrared absorption spectrum of uranyl oxalate (Nujol mull method)

Wave numbers in cm⁻¹

Fig. 9. Infrared absorption spectrum of uranyl salicylate (Nujol mull method)

TABLE I

Infrared absorption bands of uranyl phosphate

Wavenumber of the band in cm⁻¹			
Potassium bromide disc method	Nujol mull method	Fluorescence	Remarks
1	2	3	4
	550 (vs)		
	607 (w)		
	615 (m)		
	666 (w)		
671 (w)	680 (w)		
704 (m)	707 (m)		
	723 (w)		
	749 (s)		
818 (w)			
828 (m)	834 (s)	828	ν_1 fundamental
852 (w)			
877 (w)			
921 (vs)	926 (vs)	909	ν_3 fundamental
938 (w)			
954 (w)			
1011 (vs)	1020 (s)		$182-828=1010$
1119 (vs)	1110 (vs)		$182+921=1103$
1359 (w)			
1401 (s)			
1429 (s)			
1468 (s)			
1484 (m)	1500 (vs)		
1656 (s)			$2 \times 828=1656$
1712 (w)			
1848 (m)			$2 \times 921=1842$
	1900 (m)		
1972 (w)			
2079 (m)			
	2180 (m)		
2278 (w)			
2841 (w)			
2985 (w)			
3067 (w)			
3155 (vs)	3130 (vs)		Water of hydration
3425 (vs)			Water of hydration

w—weak; m—medium; s—strong; vs—very strong; b—broad.

TABLE II

Infrared absorption bands of uranyl oxalate

Wavenumber of the band in cm⁻¹			
Potassium bromide disc method	Nujol mull method	Fluorescence	Remarks
1	2	3	4
		257	ν_2 fundamental
	625 (w)	609	Oxalate frequency
	635 (w)		
672 (w)	675 (w)		
710 (w)			
723 (s)	725 (m)		
755 (w)			$3 \times 257 = 771$
796 (w)			
803 (s)	806 (s)		
849 (w)	870 (m)	875	ν_1 fundamental
928 (vs)			
947 (s)	961 (vs)	961	ν_3 fundamental
1123 (w)	1130 (w)		$849 + 257 = 1106$
1259 (s)	1250 (s)		
1316 (m)			
1355 (m)			$849 + 2 \times 257 = 1363$
1383 (w)			
1473 (w)			
1606 (s)			
1629 (vs)		1672	$C = 0$ frequency
1686 (s)	1700 (s)		
1920 (m)	1950 (w)		$2 \times 947 = 1894$
2907 (w)			
	3210 (s)		
3378 (s)			Water of hydration
3546 (w)			

TABLE III

Infrared absorption bands of uranyl salicylate

Wavenumber of the band in cm⁻¹		
Potassium bromide disc method	Nujol mull method	Remarks
1	2	3
	530 (s)	
	557 (w)	
	580 (s)	
	597 (w)	
	616 (w)	
	652 (w)	

TABLE III (*contd.*)

Infrared absorption bands of uranyl salicylate

Wavenumber of the band in cm⁻¹		
Potassium bromide disc method	Nujol mull method	Remarks
1	2	3
669 (w)	675 (m)	
695 (b)	700 (s)	
756 (vs)	755 (vs)	
803 (m)		
827 (m)	819 (s)	
848 (w)		
859 (w)		
875 (m)	870 (s)	ν_1 fundamental
892 (m)		
929 (vs)	945 (vs)	ν_3 fundamental
1033 (w)	1035 (w)	
1101 (m)		
1147 (m)		
1160 (w)		
1219 (m)		
1242 (m)	1250 (vs)	
1309 (w)		
1319 (w)		
1340 (m)		
1385 (vs)		
1393 (w)		
1420 (m)	1440 (vs)	
1466 (vs)		
1488 (m)		
1517 (m)		
1546 (m)		
1597 (w)		
1616 (s)	1600 (m)	C=0 frequency
1653 (w)		
1805 (w)		
1949 (w)		
	2800 (s)	
3215 (m)		
3367 (s)		Water of hydration

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SINGLET→TRIPLET ABSORPTION IN HALOGEN SUBSTITUTED TOLUENES*

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ABSTRACT. Continuous singlet→triplet absorption in the near ultraviolet region due to orthobromotoluene, parachlorotoluene and metafluorotoluene in the liquid state has been investigated. It has been observed that the region of absorption shifts gradually towards the red with the increase in the atomic weight of the substituent halogen atom. The luminescence spectrum due to metafluorotoluene in the solid state at −180°C has also been studied, and the luminescence bands have been found to be weaker than those due to chloro- and bromo-toluene.

INTRODUCTION

It is well known that Lewis and Kasha (1944) first suggested that the phosphorescence exhibited by many pure substances are due to transitions from the triplet state of the molecules to the lowest singlet state. Lewis and Kasha (1945) also observed singlet→triplet (S→T) absorption in pure para dichlorobenzene, dibenzalacetone, nitrobenzene and phenazine. Reid (1950a) next observed (S→T) absorption bands in pyridine in the liquid state at room temperature and in ethylene in the vapour state (Reid, 1950b). Later, Kasha (1952) demonstrated that the singlet→triplet absorption in aromatic compounds is strengthened not only by heavy substituent atoms in the molecule but also by such atoms in surrounding solvent molecules in any solution. McClure *et al.* (1954) observed S→T absorption in para diiodobenzene, para dibromobenzene, bromobenzene, β-iodochloro-and β-iodobromo naphthalenes, 1, 3, 5-tribromobenzene and 1, 2, 4, 5-tetrabromobenzene, but they did not find any such absorption in para dichlorobenzene, para iodobiphenyl and nitrobenzene either at the room temperature or at liquid nitrogen temperature. The apparent non-appearence of any S-T bands in the above cases was attributed by them to the difficulties of separating the very weak and diffuse S→T absorption spectra from the normal singlet→singlet absorption. Biswas (1954, 1955a & b, 1956a & b) and Sirkar and Biswas (1956) observed luminescence bands in some halogen substituted toluenes and although Biswas (1956) looked for the absorption band responsible for the emission he was

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*Communicated by Prof. S. C. Sirkar

unable to detect any strong S→T absorption. Recently, it has been shown by the present author (Roy, 1959) that the luminescence in parachlorotoluene in the solid state at −180°C takes place after direct S−T transition.

It was thought worthwhile to investigate whether any singlet→triplet absorption is exhibited by these compounds and whether the intensity of such absorption depends on the atomic weight of the heavy substituent atom in the molecule as reported by previous workers in other cases (McClure, *et al.* 1954). In the present investigation the singlet→triplet absorption in metafluorotoluene, parachlorotoluene and ortho bromotoluene have been studied using a long absorbing path.

The luminescence spectrum of pure metafluorotoluene in the solid state at −180°C has also been investigated and it has been compared with the spectra due to chlorotoluenes and bromotoluenes reported by previous workers (Biswas, 1956a & b).

<center>EXPERIMENTAL</center>

The liquids orthobromotoluene, parachlorotoluene and metafluorotoluene of chemically pure quality supplied by British Drug House (England), were distilled several times under reduced pressure and fractionated to remove traces of impurities. For studying the absorption spectra of the above substances in the liquid state at 24°C, a 10 cm. long pyrex glass cell provided with plane parallel quartz windows was used. A 250 watt straight filament lamp in glass envelope was employed as the source of continuous radiation in the near ultraviolet region. The absorption spectra of the substances were photographed on Ilford HP3 films using a Hilger medium quartz spectrograph having a dispersion of 22 Å/mm in the 3500Å region. The time of exposure varied from a few seconds to half a minute and the width of the slit was about 0.10 mm. Iron arc spectrum was recorded on each spectrogram as comparison. The absorption spectrum of benzene was also recorded for comparing it with the spectra of the disubstituted benzenes mentioned above. The microphotometric records of the spectrograms were obtained with a self-recording Moll microphotometer supplied by Kipp and Zonen. The absorption spectra were calibrated with the help of the microphotometric records of the iron lines after drawing a thin and straight reference line across the spectra, coinciding with a known iron line.

The luminescence spectrum of pure meta fluorotoluene in the solid state at −180°C was investigated with the arrangements used previously by Biswas (1956a).

<center>RESULTS AND DISCUSSION</center>

The microphotometric records of the absorption spectra due to pure orthobromotoluene, parachlorotoluene, metafluorotoluene and benzene are reproduced

in Figs. 1(a), 1(b), 1(c) and 1(d) respectively. The reference line in the records is the 4046 Å line of mercury.

The main bands in the luminescence spectrum due to metafluorotoluene in the solid state at −180°C with their widths and estimated intensities (s—strong, m—medium, w—weak, etc.) are given in Table I.
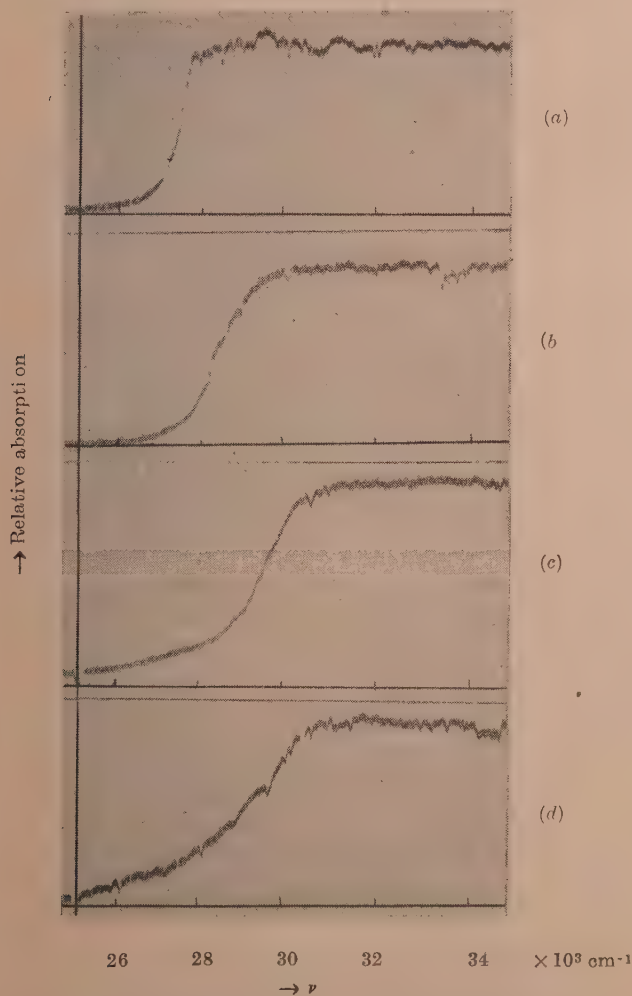

Fig. 1. (a) o-Bromotoluene. (b) p-Chlorotoluene
(c) m-Fluorotoluene, (d) Benzene

6

TABLE I

Luminescence spectra of metafluorotoluene at −180°C.

Position of the bands ν in cm^{-1}	Width of the bands in cm^{-1}.	Differences from the 1st. band in cm^{-1}
24968 (w)	436	
24072 (w)	725	896
23413 (s)	603	1555
22521 (m)	558	2447
21761 (s)	948	3207
20910 (vw)	765	4058

DISCUSSION

(a) *Absorption spectra due to singlet → triplet transition.*

It is well known (Lewis and Kasha, 1945) that benzene shows a weak S→T absorption in the region (3300-3700)Å. This is evident from Fig. 1(d). The absorption due to metafluorotoluene reproduced in Fig. 1(c) appears to be similar to that due to benzene (Fig. 1(d)). This similarity indicates the existence of very weak S→T absorption in metafluorotoluene. Fig. 1(b) and 1(a) show that as the fluorine atom is replaced successively by chlorine and bromine atoms, the S→T absorption becomes much stronger and the region where the absorption starts shifts gradually towards red. Further, absorption curves due to orthobromotoluene and parachlorotoluene show steeper rise in this region than in the case of either metafluorotoluene or benzene. These results are similar to those reported by McClure *et al.* (1954) for substituted napthalenes. It is evident from results of the present investigation that in the case of substituted benzenes also the perturbation due to substituent halogen atom increases with the increase in the atomic weight of the substituent atom and the S→T transition becomes allowed.

The strong S→T absorption observed in the cases of orthobromotoluene and parachlorotoluene also explains the intense luminescence in the visible region exhibited by these compounds in the solid state at −180°C (Biswas, 1956a, 1956b).

(b) *Luminescence spectrum of metafluorotoluene.*

It would be interesting to compare the intensities of luminescence spectrum of metafluorotoluene with that due to orthobromotoluene or parachlorotoluene excited under similar conditions. It can be seen from the results obtained by Biswas (1956 a & b) that the intensity of luminescence spectrum of orthobromo-

toluene is larger than that due to parachlorotoluene. The intensity of the lumine-scence spectrum due to metafluorotoluene recorded under similar conditions has been found to be lower than that due to para chlorotoluene. This agrees with the fact that the strength of S→T absorption is small in this case and shows further that the luminescence is produced by the transition from the triplet to the singlet state.

ACKNOWLEDGMENT

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Letters to the Editor

11

LIGHT ABSORPTION IN PARAMAGNETIC Co++ IONS IN STATE OF SOLUTION

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(*Received, May 7, 1960*)

In a recent paper Chakravarty and Chatterji (1959) assuming an approximate tetragonal axis of symmetry for the octahedron of water cluster about Co++ ion·and following the method of Abragram and Pryce (1951) have developed general expressions for the magnetic susceptibilities along and normal to the axis of symmetry of the water cluster about Co++ ion in crystal in terms of the tetragonal splitting Δ, the effective Lande-splitting factor and the spin-orbit coupling coefficient. In order to get a fit with the experimentally observed data of magnetic susceptibility measurements by Bose (1948), they required a small value of Δ varying with temperature and a value of the coefficient of spin-orbit coupling which is the same as that for the free ion, for the salts studied.

An examination of the energy level expressions Owen (1955) shows that if Δ is small, then the number of absorption bends of Co++ ion salts in crystalline state or in state of solution will be limited to three instead of six. Absorption measurements of about fifteen cobalt salts in state of aqueous solution by us, show a single band at about 19,550 cm⁻¹ and two discernable bands at about 20,900 cm⁻¹ and 21,800 cm⁻¹ which point out clearly that Δ should be extremely small in them, in agreement with the theoretical findings of Chakravarty and Chatterji. The second findings by these authors that the spin-orbit coupling coefficient in crystalline state is the same as the free ion value, will mean that the covalent overlap of 3d-orbitals and the s-and p-orbitals of water cluster about Co++ ion can be neglected (Owen 1955). Consequently the term separation E for the free ion will be the same as E', that for the crystal. From our optical absorption

336

measurements we have evaluated E' for about a dozen of Co^{++} salts in state of aqueous solution. We find that $E'/E \sim 0.95$ which is in complete agreement with the findings of Chakravarty and Chatterji.

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